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Title: A Technical basis for in-house calibration of 252Cf neutron source

emission rates

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Introduction:

10CFR835 stipulates that radiation protection instruments must be calibrated at least annually. Accordingly, calibration of dose rate instruments are reliant on well-known reference fields. The neutron-free-in-air facility (NFIA) located at TA36-0214 provides such a capability for neutron remmeters. One of the reference NFIA sources, 252 Cf, must be replaced every 8-10 years due to its relatively short half-life (2.645 \pm 0.008 y). In the past, each newly purchased 252 Cf source has been calibrated at NIST using the Mn-bath technique prior to shipping to LANL. However, because of COVID-19 complications, the most recently acquired 252 Cf source (FTC-CF-7167) has been stored at LANL pending approval to ship to NIST for calibration. Due to the considerable expense in transporting the source to and from NIST, this TBD was written to demonstrate that new sources can be accurately calibrated via intercomparison measurements with older NIST-calibrated 252 Cf sources. It had been previously noted that such measurements yielded emission rates that agreed very well with the official rates established by NIST.

In principle any neutron-sensitive instrument can be used for intercomparison purposes as long as the measurements are made under identical conditions. This TBD focuses on two specific neutron instruments; ROSPEC and SWENDI.

ROSPEC (a <u>ro</u>tating neutron <u>spec</u>trometer) is comprised of six gas proportional counters that when combined provide spectroscopic data from thermal energies to 5 MeV. Each counter has an associated 256 channel pulse height spectrum. The data reported here was obtained with a ROSPEC (S/N 0002) which has demonstrated remarkable stability over the past 20 years as illustrated, for example, by Fig. 1 where pulse height data for the 10atm H₂ counter is plotted normalized for run time and neutron emission rate. The data shown in Fig.1 were recorded for various bare Cf sources at 100cm¹.

SWENDI is a cylindrical polyethylene-moderated rem meter based on a 2 atm ³He thermal neutron counter. The inherent pulse height discrimination property of ³He ensures that gamma interference is negligible. SWENDI also has a relatively high sensitivity on the order of 450 cpm per mrem/h for bare ²⁵²Cf.

Previous supporting measurements:

Prior "unofficial" ROSPEC calibration measurements have strongly suggested that an in-house determination of emission rate can be done accurately. As demonstrated, for example, by the data shown in Table 1 which summarizes an analysis of nine ROSPEC runs done over the course of 18 years for three different bare Cf sources at a common distance of 100cm. Table 1 presents the average count rates (cps) of four counters over their respective neutron region-of-interests

¹ where, for example, May29A02 stands for a run on 5/29/2002 and "A" indicates the first run of the day

(ROI) after normalizing for run time and the decay-corrected 252 Cf neutron emission rate (as based on the source's NIST calibration certificate) on the date of the run. Aside from these average responses (ROI cps per n/s), Table 1 also lists an estimated uncertainty for each counter. These uncertainties were based strictly on an unweighted average of the nine individual run results without regard to the underlying number of ROI counts (which were typically >> 1E04).

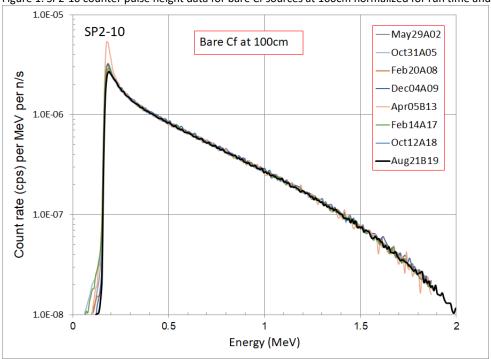


Figure 1. SP2-10 counter pulse height data for bare Cf sources at 100cm normalized for run time and source emission rate

The average conversion coefficients (ROI cps per n/s) for each counter (Table 1) were then applied to their respective counter in the nine bare Cf runs. Then the individual estimated neutron emission rates gained from each counter were averaged to give an estimated neutron emission rate for each run. Finally, these estimated values were decay-corrected ($t_{1/2}$ = 2.645y) to the reference date on the applicable source's NIST calibration certificate. Table 2 compares

Table 1. ROSPEC counter response for bare Cf sources @ 100cm on NFIA

ROSPEC counter ID	ROI (MeV)	ROI cps per n/s	% uncertainty
SP2-1 (0.75 atm. H ₂ gas)	0.050 - 0.225	5.57E-08	2.9
SP2-4 (4 atm. H ₂ gas)	0.150 - 0.750	2.68E-07	2.9
SP2-10 (10 atm. H ₂ gas)	0.400 - 1.50	4.22E-07	2.6
SP6 (5 atm. P10 gas)	1.25 - 5.00	3.88E-07	3.5

the source emission rates determined by NIST and ROSPEC. Where, for example, two of the nine runs involved Cf source FTC-CF-777. The last column in Table 2 averages the individual run estimates for each Cf source which are seen to be in remarkably good agreement with the emission rates measured at NIST.

Using this "standalone" measurement technique, a single ROSPEC run on 9/3/2020 with just the SP2-10 counter² yielded an emission rate of 6.88E08n/s for the new Cf source (FTC-CF-7167) as shown in Table 2.

Table 2. Comparison of ROSPEC-based ²⁵² Cf emission rates with NIST calibrated values. Uncertainties are 2 σ v	e 2σ values
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Source ID	Reference	NIST calibration (n/s)	LANL "calibration" (n/s) on ref. d	
	date		Individual runs	Average
FTC-CF-777	8/28/1996	3.667E08 ± 2.90 %	3.61E08, 3.47E08	3.54E08
FTC-CF-1899	5/19/2003	3.520E08 ± 4.30 %	3.54E08, 3.66E08,	2 50500
F1C-CF-1899	5/19/2003	3.520EU8 ± 4.30 %	3.61E08, 3.50E08	3.58E08
FTC-CF-Z3899	5/17/2010	9.530E08 ± 2.60%	9.45E08, 9.65E08,	9.55E08
F1C-CF-23699	3/1//2010	9.550E06 ± 2.00%	9.55E08	9.55506
FTC-CF-7167	9/03/2020	2/2	6.88E08 (SP2-10	6 00500
F1C-CF-/16/	9/03/2020	n/a	only)	6.88E08

An Intercomparison method of calibrating source emission rate:

The excellent agreement with the NIST-calibrated emission rates, gave confidence that ROSPEC could accurately determine source emission rates based on standalone measurements and a source decay correction. Note that in some cases, the ROSPEC measurements listed in Table 2 were made more than 9y following the NIST calibration.

But a more accurate estimate – one that does not rely on long-term ROSPEC stability - of the emission rate of a new source can be made via an intercomparison measurement with a previously NIST-calibrated older source. On 9/3/2020, ROSPEC measurements with the new Cf source (FTC-CF-7167) and an older source (FTC-CF-Z8399) were made under identical conditions at a distance of 100cm. Due to the relatively high emission rate of the new source and the count rate limitations of ROSPEC only the SP2-10 counter was enabled for the intercomparison runs.

Table 3 summarizes the data collected for the SP2-10 counter. In order to eliminate any possibility of gamma interference, the low energy boundary of the ROI was increased to 0.5 MeV (channel 80).

²The count rate with all counters active was too high (>1600 cps)

Table 3. Results of intercomparison measurements with ROSPEC (SP2-10 counter) of old and new Cf sources

Source ID	t(s)	ROI counts	ROI cps	Elapsed time since	Count rate ratio
				NIST calibration (y)	
FTC-Cf-Z3899	1650	32948	19.968	10.299	10 022 ± 0 600/
FTC-Cf-7167	300	64827	216.09	n/a	10.822 ± 0.68%

The emission rate of the older source was estimated from Eq. 1³ which considered not only the decay of ²⁵²Cf but also the contributions of ²⁵⁰Cf and ²⁴⁸Cm both of which become increasingly more significant with time. The initial (t=0) emission rate of ²⁵⁰Cf was estimated as 0.1% of the ²⁵²Cf rate based on a typical ²⁵⁰Cf atomic fraction of 0.17. Strictly speaking, t=0 corresponds to the date the Cf isotopes were last chemically separated prior to source preparation. If this information is available along with the isotopic composition of the Cf source it should be used in the analysis that follows.

$$Q(n/s) = Q_{252}e^{-\lambda_{252}t} + Q_{250}e^{-\lambda_{250}t} + T(1 - e^{-\lambda_{252}t})$$
 Eq. 1

In Eq. 1, T is the 248 Cm emission rate when 252 Cf has fully decayed (i.e. 39.8 n/s per µg of 252 Cf) and Q, t and λ have their usual meanings. Based on the NIST-calibrated emission rate of source FTC-CF-Z3899, the initial mass of 252 Cf was calculated as $412\mu g^4$ and therefore T= 1.64E04 n/s.

Table 4 presents the half-life and decay constant values for the Cf isotopes of interest. These values and their uncertainties were taken directly or derived from data on the National Nuclear data center website, www.ndcc.gov. As the half-life of 248 Cm is 3.48E05y, its decay was neglected in the calculations to follow.

Table 4. Decay data for Cf isotopes

Isotope	t _{1/2} (y)	λ (y ⁻¹)	% unc.	
²⁵² Cf	2.645	0.2621	0.30	
²⁵⁰ Cf	13.08	0.0530	0.69	

Using Eq. 1 and data drawn from the above Tables and text, the emission rate of source FTC-CF-Z8399 as of 9/3/2020 was calculated as 6.460E07 n/s. The combined uncertainty associated with this result was calculated based on Eq. 2 where the partial derivatives were taken with respect to the variables in Eq.1 and $\sigma^2(x_i)$ was the variance associated with variable x_i . Note, Eq. 2 assumes the variables are uncorrelated.

$$\sigma^2 = \sum_{i=1}^n \left(\left[\frac{\partial Q}{\partial x_i} \right]^2 \sigma^2(x_i) \right)$$
 Eq. 2

³ Roberts and Jones, RPD 126(1-4), 83, 2007

 $^{^4}$ Based on 2.31E06 n/s per μg of ^{252}Cf

Table 5 presents the uncertainty budget calculation for the neutron emission rate of FTC-CF-Z8399 as of 9/3/2020. The relative uncertainties of Q_{250} and T in Eq. 1 were assumed to be 20% and 10% respectively. However, the largest contributions to the overall uncertainty were those associated with the original NIST calibration and the 252 Cf decay constant⁵. When summed, the individual contributions gave a total variance of 9.776E11 n^2/s^2 or a standard deviation of 9.89E05 n/s (1.53% of the decay-corrected value of 6.460E07 n/s).

Finally, the emission rate of the new source (FTC-CF-7167) as of 9/03/2020 was calculated according to Eq. 3,

$$Q(^{n}/_{s}) = 10.822 \pm 0.68\% * 6.460E07 \pm 1.53\% = 6.991E08 \pm 1.67\%$$
 Eq. 3

Table 5. Uncertainty budget for source FTC-CF-Z3899 as of 9/03/2020

variable (x _i)	9G/9xi	(∂Q/∂x _i) ²	σ_{i}^{2}	$(\partial Q/\partial x_i)^2 * \sigma_i^2$
Q ₂₅₂	exp(-λ ₂₅₂ t)	4.524E-03	1.53E14 n ² /s ²	6.944E11 n ² /s ²
Q ₂₅₀	exp(-λ ₂₅₀ t)	0.2623	$3.56E10 \text{ n}^2/\text{s}^2$	9.338E09 n ² /s ²
λ_{252}	$t(1 - exp(-\lambda_{252}t)) (T-Q_{252})$	$4.359E17 n^2 y^2/s^2$	6.28E-07 y ⁻²	2.738E11 n ² /s ²
λ_{250}	t(1 - exp(-λ ₂₅₂ t)) Q ₂₅₀	$3.710E13 n^2 y^2/s^2$	1.33E-07 y ⁻²	4.214E06 n ² /s ²
Т	$(1 - \exp(-\lambda_{252}t))$	0.8700	$2.63E06 \text{ n}^2/\text{s}^2$	2.287E06 n ² /s ²

Using a coverage factor of 2 (i.e. a 95% confidence interval), the expanded uncertainty of source FTC-CF-7167 as of 9/3/2020 was calculated as 3.35%. Note that the emission rate determined using the standalone SP2-10 measurement (6.88E08 n/s in Table 2) falls within this expanded uncertainty.

An EXCEL spreadsheet (*Calibration of Cf emission rate and uncertainty.xlsx*) has been setup to facilitate the above calculations and a copy has been placed on the RP drive in the $\RP-SVS\ RIC\$ *Cf_calibration* folder.

<u>Intercomparison exercise with older NIST-calibrated bare Cf sources</u>

On 11/20/2020 another set of intercomparison runs were conducted in the same manner as those described above. But this series of runs included older NIST-calibrated Cf sources to see how far back in time the intercomparison method could be used and still be able to accurately and precisely establish the emission rate of a newly acquired source.

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⁵ At least for elapsed times of <10y

Table 6 details the NFIA Cf sources available as of 11/20/2020 where Eq.2 was again applied to yield the uncertainty in emission rates. Table 7 presents the results of the intercomparison runs at a fixed distance of 100cm relative to the center of ROSPEC. Note that ROSPEC was operated in rotational mode and that only the SP2-10 counter ROI (channels 80-200) count rates were compared. It was not necessary that ROSPEC be rotating (i.e. it could have been operated while stationary) though it was, of course, imperative that all runs were made using the same operating mode.

Table 6. Emission rates of Cf sources in use at the NFIA facility as of 11/20/2020

	NIS	ST calibration	As of 11,	/20/2020	
Source ID	Reference date	$Q_{ref}(n/s)$	%unc (1σ)	Q(n/s)	%unc (1σ)
FTC-Cf-777	8/26/1996	3.667E08	1.45	7.47E05	3.39
FTC-Cf-1899	5/19/2003	3.52E08	2.15	3.72E06	2.57
FTC-Cf-Z3899	5/17/2010	9.53E08	1.30	6.11E07	1.55
FTC-Cf-7167	n/a	n/a	n/a		

Table 7. ROSPEC SP2-10 ROI count data and estimated emission rate of FTC-CF-7167 as of 11/20/2020 based on intercomparison with NIST-calibrated sources

		Gross	Cross	Ratio wrt	Gross	Calculated	%unc (1σ) in
Source ID	t(s)	ROI	Gross	FTC-Cf-	cps per	Q(n/s) for	Q(n/s) for
		counts	ROI cps	7167	n/s	FTC-Cf-7167	FTC-Cf-7167
FTC-Cf-777	9151	2083	0.2276	867.21	3.05E-07	6.48E08	4.04
FTC-Cf-1899	6250	6950	1.1120	177.50	2.99E-07	6.60E08	2.85
FTC-Cf-Z3899	3100	55803	18.001	10.965	2.94E-07	6.70E08	1.62
FTC-Cf-7167	600	118426	197.377				

SWENDI intercomparison results with older NIST-calibrated bare Cf sources

Initial intercomparison runs using SWENDI⁶ were done on 12/19/2019 that compared the count rates for the NIST-calibrated FTC-CF-Z3899 Cf source with the new FTC-CF-7167 source. These runs were done at several distances ranging from 50 to 295 cm for each source. On 12/19/2019, the emission rate of FTC-CF-Z3899 was calculated as 7.77E07 n/s with an uncertainty of 1.50% (1σ).

Table 8 summarizes the SWENDI data collected on 12/19/2019 and the estimated emission rate of FTC-CF-7167 on that date. All data was collected using three consecutive 120s runs for each source and distance. Based on the count rate ratios and the known FTC-CF-Z3899 emission rate

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⁶ SWENDI ESH# 13682 with E-600 ESH# 12026

and associated uncertainty, the emission rate and its uncertainty for FTC-CF-7167 were calculated at each measurement distance as shown in Table 8. A weighted average of the data shown in this Table (excluding the 50cm data) gave an estimated emission rate for Cf source FTC-CF-7167 of 8.70E08 n/s with an uncertainty of 1.51%.

Table 8. Summary of SWENDI intercomparison runs on 12/19/2019 and resulting estimated emission rate for FTC-CF-7167

d(cm)	FTC-CF-Z3899	FTC-CF-7167	Count rate ratio	Calculated	Uncertainty (1σ)
	total counts	total counts	and uncertainty	Q(n/s) for	in Q(n/s) for
				FTC-CF-7167	FTC-CF-7167
50	1112648	11968762	10.751 ±0.10%	8.36E08	1.50%
100	278476	3118312	11.198 ±0.20%	8.70E08	1.51%
150	130736	1470564	11.248 ±0.29%	8.74E08	1.53%
200	79822	889732	11.146 ±0.37%	8.66E08	1.54%
250	55260	620228	11.224 ±0.44%	8.72E08	1.56%
295	43084	481376	11.173 ±0.50%	8.68E08	1.58%

Almost a year later, on 11/3-4/2020, a series of similar intercomparison runs with all the previously NIST-calibrated Cf sources and FTC-CF-7167 were done using the same SWENDI/E600 instruments used previously. These bare Cf runs were again done at various distances from each source but only those at 100cm will be discussed here.

Table 9 presents the SWENDI data (averaged over 3 consecutive runs of the same duration for each source). Note that SWENDI count rates are about 35 times higher than observed for the ROSPEC's SP2-10 counter (Table 7) with the same Cf source a couple of weeks later (i.e. 11/20/2020). Based on the SWENDI intercomparison data, estimated emission rates for the new source were calculated as shown in Table 9. The estimated emission rates for FTC-CF-7167 are seen to be remarkably consistent which suggests that intercomparison measurements

Table 9. SWENDI count rate data and estimated emission rate of FTC-Cf-7167 as of 11/3/2020 based on relative count rates with NIST-calibrated sources.

		Cross	Cross	Ratio wrt	Gross	Calculated	%unc (1σ)
Source ID	t(s)	Gross	Gross	FTC-Cf-	cpm per	Q(n/s) for –	in Q(n/s) for
		counts	cpm	7167	n/s	FTC-CF-7167	FTC-CF-7167
FTC-Cf-777	3x600	13320	444	941.33	5.89E-04	7.11E08	3.48
FTC-Cf-1899	3x300	34290	2286	182.83	6.06E-04	6.88E08	2.62
FTC-Cf-Z3899	3x120	224706	37451	11.169	6.05E-04	6.90E08	1.55
FTC-Cf-7167	3x120	2507694	417949				

against sources calibrated more than 24 years ago (i.e. in 1996) can provide accurate and sufficiently precise estimates of the emission rates of new sources. The proviso being that proper account of the time-dependent emission rate of the older source must be taken into consideration.

The 12/19/2019 estimate of the emission rate of FTC-CF-7167 (Table 8) yielded a value of 6.92E08 n/s ($\pm 1.51\%$) when decay-corrected to 11/3/2020 in excellent agreement with the data given in Table 9.

<u>Discussion of bare Cf intercomparison runs</u>

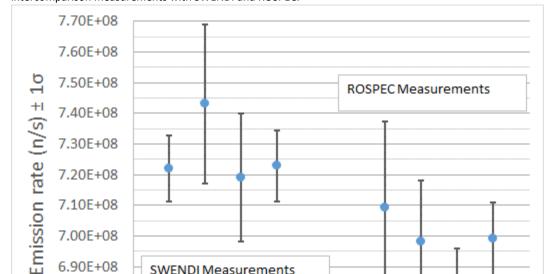
The intercomparison runs discussed above were done over a period of several months. To aid comparison of the various estimates of the FTC-CF-7167 neutron emission rate, the above data (Tables 7,8 and 9) were decay-corrected to a common date of 9/03/2020. The results are summarized in Table 10 and shown in graphical form in Figure 2.

Table 10. Decay-corrected estimates of the FTC-CF-7167 neutron emission rate as of 9/03/2020 based on all intercomparison measurements with bare Cf sources.

measurements with ba	ie Ci sources.				
Date of	NIST-calibrated	Technique	Instrument	Q(n/s) as of 9/03/2020	% unc.
measurement	source ID			9/03/2020	
12/19/2019	FTC-Cf-Z3899	Bare Cf	SWENDI/E-600	7.22E08	1.5
9/03/2020	FTC-Cf-Z3899	Bare Cf	ROSPEC (SP2-10)	6.99E08	1.7
11/03/2020	FTC-Cf-777	Bare Cf	SWENDI/E-600	7.43E08	3.5
11/04/2020	FTC-Cf-1899	Bare Cf	SWENDI/E-600	7.19E08	2.9
11/04/2020	FTC-Cf-Z3899	Bare Cf	SWENDI/E-600	7.23E08	1.6
11/20/2020	FTC-Cf-777	Bare Cf	ROSPEC (SP2-10)	7.09E08	4.0
11/20/2020	FTC-Cf-1899	Bare Cf	ROSPEC (SP2-10)	6.98E08	2.9
11/20/2020	FTC-Cf-Z3899	Bare Cf	ROSPEC (SP2-10)	6.85E08	1.6

Figure 2 clearly shows that the emission rate estimates determined by each instrument are statistically equivalent. Table 11 gives the weighted average and associated uncertainty of the emission rate of FTC-CF-7167 as of 9/3/2020 as yielded by each instrument as well as their combined estimate.

The combined estimate of 7.10E08 $\pm 0.72\%$ n/s for the emission rate of FCT-CF-7167 as of 9/03/2020 was more than adequate for characterizing the Cf-based NFIA reference fields. Nevertheless, the ~4% difference in the SWENDI and ROSPEC estimates (Table 11) was cause of some concern and follow up measurements were made in an attempt to resolve.



SWENDI Measurements

6.90E+08

6.80E+08

6.70E+08

Figure 2. Estimated neutron emission rate of FTC-CF-7167 and associated uncertainty ($\pm 1\sigma$) as of 9/3/2020 based on intercomparison measurements with SWENDI and ROSPEC.

Table 11. Summary of neutron emission rate of FTC-CF-7167 as 9/03/2020 based on bare Cf intercomparison measurements

Instrument	Q(n/s)	Relative uncertainty (1σ)
SWENDI	7.24E08	0.98%
ROSPEC	6.94E08	1.04%
ROSPEC + SWENDI	7.10E08	0.72%

One possible explanation for the ~4% bias in the SWENDI and ROSPEC results was the spectral sensitivity of each instrument. ROSPEC relied on a relatively narrow ROI i.e. recoil protons between approximately 0.5 and 1.5 MeV while SWENDI has a continuous response across the entire spectral envelope. When coupled with slight differences in the encapsulation from one source to another (i.e. anisotropy effects), it's not unexpected that each instrument would respond differently (to both direct and room-scattered neutrons) and possibly account for the bias noted in the intercomparison runs with bare Cf sources.

At NIST, a Mn-bath calibration technique is used to determine neutron emission rates. The method is based on comparing the thermal neutron activation rates of ⁵⁵Mn of an unknown source against a source of well-known emission rate. The Mn bath efficiently thermalizes source neutrons and, as it's essentially a 4π counting geometry, passively compensates for any source anisotropy. With this in mind, a final series of intercomparison runs were done using the NFIA's 30cm-diameter Cd-clad D₂O moderator to randomize any directional dependence of neutrons (emission rate and/or spectral) emitted by the various Cf sources.

Intercomparison using D2O-moderated Cf sources

On 12/22/2020, a series of intercomparison runs were done in an attempt to resolve the SWENDI vs ROSPEC bias observed in the earlier data. To this end, ROSPEC and SWENDI data was collected using D_2O -moderated Cf sources. These measurements were again conducted at a fixed distance of 100cm from the source. Table 12 lists the neutron emission rate data for the NIST-calibrated NFIA Cf sources on 12/22/2020.

Table 12. Emission rate data for the NFIA Cf sources as of 12/22/2020

	NIS	ST calibration	As of 12	/22/2020	
Source ID	Reference date	$Q_{ref}(n/s)$	%unc (1σ)	Q(n/s)	%unc (1σ)
FTC-Cf-777	8/26/1996	3.667E08	1.45	7.32E05	3.42
FTC-Cf-1899	5/19/2003	3.52E08	2.15	3.64E06	2.57
FTC-Cf-Z3899	5/17/2010	9.53E08	1.30	5.97E07	1.54
FTC-Cf-7167	n/a	n/a	n/a		

This series of intercomparison runs were also an opportunity to consider ROSPEC counters other than the SP2-10. The reduced count rates in the D_2O fields (about a factor of 5 relative to bare Cf) enabled the simultaneous use of other counters without exceeding the instrument's maximum count rate. So, in addition to the SP2-10 counter, the SP2-1 and SP2-4 counters (Table 1) were also activated for the D_2O -moderated Cf runs.

These runs were also an opportunity to carefully verify that the ROSPEC (and SWENDI) data was collected under equilibrium count rate conditions. It is well-known that the count rates from both instruments will increase slightly (~2-5%) over the course of a prolonged exposure before stabilizing. ROSPEC stability was verified by sandwiching consecutive runs with the new source (FTC-CF-7167) with a longer run using the FTC-CF-Z3899 NIST-calibrated source. The consistency in the initial and final count rates with the newer source was taken as evidence that count rate equilibrium had been established. Only the FTC-CF-Z3899 Cf source was used for the ROSPEC measurements on 12/22/2020.

In the case of ROSPEC, these measurements also considered the small but finite background count rate in each of the counters. ROSPEC counters are subject to random internal arcs and/ or sparks that generate counts in the ROI channels. The count rates have a dependence on operating voltage and, for instance, in the case of the SP2-10 counter (4000v) can vary between 20-60 cph. Typically, the background rate is highest after a long period of nonuse but once high voltage is reapplied, steadily decreases with continuous operation.

Table 13 gives the background count rates for the three ROSPEC counters used in the 12/22/2020 measurements. The background data, recorded just prior to the D_2O

measurements, was taken over channels 80-200 in each counter to eliminate any gamma-induced counts. The background rates in Table 13 are representative of typically observed values.

Table 13. Background count rates in ROSPEC counters.

		SP2-1			SP2-4		SP2-10			
t(s)	counts	cps	%unc	counts	cps	%unc	counts	cps	%unc	
7977	3	0.0004	0.02%	162	0.020	0.16%	114	0.014	0.13	

Table 14 presents the gross count rate data for the ROSPEC counters collected during the D_2O -moderated source exposures. In each case, a common ROI (channels 80-200) was again used after establishing that count rate stability had been reached.

Table 14. Gross counts and count rates of ROSPEC counters for D₂O-moderated old and new Cf sources.

		SP2-1				SP2-4		SP2-10		
Cf source	t(s)	gross counts	gross cps	%unc	gross counts	gross cps	%unc	gross counts	gross cps	%unc
FTC-CF-7167	600	5962	9.937	3.03%	13651	22.752	2.01%	22379	37.298	1.55%
FTC-Cf-Z3899	1516	1337	0.882		3037	2.003		5148	3.396	
FTC-Cf-7167	600	5980	9.967	3.03%	13815	23.025	2.00%	22486	37.477	1.55%

The net count rate data for each ROSPEC counter is given in Table 15. In addition, the count rate ratios of the new and older Cf sources are also given along with their relative standard deviation.

		SP2-1				SP2-4				SP2-10			
		net	net			net	net			net	net		
Cf source	t(s)	counts	cps	ratio	%unc	counts	cps	ratio	%unc	counts	cps	ratio	%unc
FTC-CF-7167	600	5962	9.936	11.27	3.03%	13639	22.731	11.46	2.02%	22370	37.284	11.03	1.56%
FTC-Cf-Z3899	1516	1336	0.882			3006	1.983			5126	3.381		
FTC-Cf-7167	600	5980	9.966	11.31	3.03%	13803	23.005	11.60	2.02%	22477	37.462	11.08	1.56%

The weighted average of the six individual net count rate ratios was determined as $11.230 \pm 0.81\%$. As of 12/22/2020, the emission rate of the NIST-calibrated FTC-CF-Z3899 source was $5.97E07 \pm 1.54\%$ n/s (Table12) from which the estimated emission rate of FTC-CF-7167 on 12/22/2020 was calculated as $6.70E08 \pm 1.74\%$ n/s. When decay-corrected, the emission rate of

FTC-CF-7167 on 9/3/2020 was determined to be $7.25E08 \pm 1.74\%$ n/s. This result is in excellent agreement with the value estimated based on the SWENDI intercomparison measurements with bare Cf sources (Table 11). This was due to establishing count rate stability, accounting for background count rates and including the slightly higher emission rates derived from the SP2-1 and SP2-4 counter data.

SWENDI data

The SWENDI/E-600 instrument combination used previously for the bare Cf runs was again used for the D₂O-Cf intercomparison measurements. Like the ROSPEC counters, SWENDI had a lower count rate in the relatively low energy D₂O-Cf reference fields than for bare Cf as demonstrated by the data shown in Table 16. Even so, statistically significant data was obtained in a reasonably short period of time even for the weaker Cf sources. The background count rate was <1 cpm so no correction for background was warranted.

As done previously, the SWENDI data was collected using one or more consecutive scalar counts at a distance of 100cm. Table 16 summarizes the SWENDI data obtained (under count rate equilibrium) for three NIST-calibrated Cf sources as well as for the most recently acquired source (FTC-CF-7167). The count rate ratios (new source/older source) are seen to be in excellent agreement with those observed during the bare Cf measurements (Table 9). Based on the count rate ratios and the 12/22/2020 emission rates of the calibrated sources, the estimated emission rate of the new source was calculated along with its associated relative uncertainty as shown in Table 16.

Table 16. SWENDI data in D₂O-moderated Cf fields and calculated emission rate of FTC-CF-7167 as of 12/22/2020

Source ID	t(s)	counts	cpm	Ratio wrt FTC-Cf- 7167	cpm per n/s	Calculated Q(n/s) for FTC-Cf- 7167	%unc (1σ)
FTC-Cf-777	10x300	6500	130	921	1.78E-04	6.74E08	3.63
FTC-Cf-1899	16x60	10621	663.8	180.3	1.824E-04	6.56E08	2.75
FTC-Cf-Z3899	8x60	85712	10714	11.170	1.792E-04	6.67E08	1.58
FTC-Cf-7167	420	837746	119678				

Again, as observed with the bare Cf runs the individual estimates of the emission rate of FTC-CF-7167 are in statistical agreement. The agreement with the ROSPEC intercomparison measurements with source FTC-CF-Z3899 was also remarkably good as the $^4\%$ bias was no longer evident.

Based on the four intercomparison estimates (combined ROSPEC and three SWENDI measurements) of FTC-CF-7167 emission rate, a weighted average of 6.67E08 n/s \pm 1.03% as of 12/22/2020 was calculated. When referenced to 9/3/2020, the D₂O-moderated intercomparison runs resulted in an estimated emission rate of 7.22E08 n/s \pm 1.03%. This estimate was in excellent agreement with the bare Cf intercomparison result with the added bonus of resolving the bias in the SWENDI and ROSPEC data.

Discussion

The DOE Implementation guide to 10CFR835 (DOE G 441.1C) states in section 9.6 that calibration field accuracies <u>should</u> be in accord with N323A (now N323AB). In turn, N323AB states a 10% accuracy (k=1) for neutron fields with respect to neutron dose rate should be maintained and furthermore must be (i.e. a "shall" requirement) NIST-traceable. The proposed intercomparison method easily satisfies both N323AB requirements⁷ on the assumption that 1) the conversion from fluence rate to dose rate introduces (as fully anticipated) minimal additional variance, 2) NIST traceability can be indirectly conferred and 3) any additional sources of uncertainty as outlined in RP-SVS-TP-065 are minimal.

The limited set of intercomparison measurements discussed above imply that relative count rates with Cf sources as old as 24 years have merit in establishing the emission rate of a newly acquired source - as long as the contributions of 250 Cf and 248 Cm are considered. However it may be prudent to limit intercomparisons to sources < 20y old where such considerations are minor. In practice, this would mean that a NIST calibration would only be required every other Cf source acquisition.

Based on the intercomparison measurements the following recommendations are proposed whenever a newly acquired Cf source is to be calibrated in-house:

- SWENDI is preferred over ROSPEC due to:
 - SWENDI's lower background count rate
 - SWENDI's greater sensitivity (cpm per n/s)
 - SWENDI's higher dynamic range in terms of count rate
- ROSPEC measurements
 - o Employ multiple counters if conditions (i.e. count rates) allow
 - Background count rates must be known especially when gross count rates are relatively low.
- Regardless of which instrument is used:
 - A moderator such as the D₂O sphere or a large diameter polyethylene sphere should be used.

⁷ Regardless if ROSPEC or SWENDI or an average of both methods are used to calibrate a new source.

- o Count rate stability must be established prior to collecting useful data
- o Measurements are conducted at a distance of at least 100cm from the source
- o Intercomparison measurements must be done under identical conditions
- o Intercomparison data must be adequately documented and recorded

Conclusion

Subject to the above criteria, the feasibility of an in-house method of determining the neutron emission rate of a newly acquired Cf source has been demonstrated. An in-house calibration saves the cost of shipping a source to and from NIST as well as the expense of the calibration itself. Furthermore, an in-house calibration can be done within a day while a NIST calibration can take several weeks or even months. NIST calibrations will still be required but only for every other new Cf source or, even, for every third source and still be incompliance with N323AB.